Beta barium borate ($\beta$-BaB$_2$O$_4$ or BBO) has made a tremendous impact in the field of harmonic generation as a nonlinear optical material. It has proven very effective in terms of frequency doubling and for the design of highly tunable optical parametric oscillators [1]. The scope of BBO research, however, has been delegated to its optical nonlinear properties in the visible and ultraviolet region. Although it has been previously reported that its transmission extends beyond 3500 nm, its optical properties for even longer wavelengths has not drawn much interest[2]. We report experimental work on the optical properties of BBO in the 0.1 to 1.1 terahertz (THz) region. Primarily, results show that this material is significantly transparent for submillimeter waves and it was demonstrated that this material exhibits birefringence in the THz range, as reflections of lower interests in THz phonon variations in past studies. These findings could prove vital in utilizing phase matching conditions to realize BBO-based THz frequency doublers and even optical parametric amplifiers.

A type I 10mm x 10 mm x 1 mm (thick) BBO crystal (Type I, $\theta$=90\(^\circ\), $\Phi$= 90\(^\circ\), Crylight Photonics, Inc.) was used for the study. The THz radiation from a bulk InAs surface excited by a mode-locked, p-polarized Ti:Sapphire laser was used, producing transient pulses centered at ~0.8 THz ranging from 0.1 THz to ~1.5 THz. The emitted predominantly p-polarized THz radiation was collimated and focused using paraboloidal mirrors to perform azimuthal dependence of the broadband THz transmission using a liquid-He cooled Silicon bolometer detector. The angular dependence of the transmission spectra (for every 5\(^\circ\) steps over 360\(^\circ\)) was measured using a far infrared fourier transform spectrometer fitted with the same bolometer. The transmission spectra were deconvolved by dividing the raw transmission data with the THz spectrum from an aperture whose diameter is equal to the BBO crystal holder aperture.

The azimuthal angle dependence of the broadband THz transmittance is shown in Fig. 1(a). The y-axis is the % transmittance from the voltage reading of the bolometer output (the reference signal reading was 50.38 mV). The two-fold dependence of its transmission properties for a 360-degree rotation implies THz birefringence. Furthermore, the data also confirms that the crystal c-axis of the BBO is parallel to the 0\(^\circ\) sample orientation in the experiment.

The plot of the THz transmittance spectra for selected azimuthal angle orientations are shown in Fig. 1(b). It is worthwhile to note that the BBO crystal has 60% transmittance at 0.35 THz for the 0\(^\circ\) orientation. This result suggests BBO as a suitable nonlinear medium for InAs surface-based THz emitters. Moreover, an azimuthal angle dependent THz transmittance at intermediate angular orientations (i.e. from 40\(^\circ\) to 60\(^\circ\) orientations and at their corresponding equivalent angular orientations), was observed at the ~0.65 THz band (or ~21.45 cm\(^{-1}\)). This is attributed to low frequency phonon modes of the [B$_3$O$_6$]$^{3-}$ rings[3]. As such, first-principles calculations of the crystalline vibrations of a BBO crystal unit cell in the THz region were performed using periodic density functional theory methods. Figure 2(a) illustrates the associated directions of the vibrational modes of the [B$_3$O$_6$]$^{3-}$ rings and the dipole derivatives (in atomic units) for each respective mode are shown in the inset of Fig. 2(b). Four distinct phonon modes were found at ~27 cm\(^{-1}\) but only the two strongest modes are shown. Figure 2(b) shows the projection of the calculated dipole moments to the y-z

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**Fig. 1.** (a) Azimuthal angle dependence of the broadband THz transmission of the BBO crystal, showing birefringence. The 0\(^\circ\) orientation coincides with the crystal’s c-axis. (b) Terahertz transmission spectra for selected azimuthal angle orientations. A strong absorption band at 0.65 THz for the 40\(^\circ\) to 60\(^\circ\) orientations is attributed to low frequency phonon modes of the [B$_3$O$_6$]$^{3-}$ rings. The BBO crystal exhibits 60% transmittance at 0.35 THz.
plane, where the x-axis (a-axis) coincides with the THz radiation propagation direction. The angular orientations of the dipole derivative projections with respect to the vertical c-axis are 46.1° and 61.9° for the 26.97 cm⁻¹ and 26.98 cm⁻¹ modes, respectively. Maximum absorption at the 0.65 THz band should then coincide with orientations parallel to these angles and in their equivalent directions, as was observed in the experiment. The discrepancy between the experimental and calculated mode frequency may be attributed to the fact that only one BBO unit cell was used in the simulation.

The frequency spacing of the Fabry-Perot fringes in Fig. 1(b) was used to calculate the THz refractive index of the sample for different angles. The azimuthal angle behavior of the calculated refractive index values are shown in the open circle trace in Fig. 2(c) and the continuous line trace is a least squares fit.

A reasonable agreement was achieved with no and ne fitting values of 2.81357 and 2.51842, respectively. A remarkably high refractive index contrast Δn value of ~0.296 was determined. These results suggest that BBO crystals may also prove useful as a nonlinear optical material in the THz region.

REFERENCES


1Osaka University
2The Graduate University for Advanced Studies
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